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PACIFIC NORTHWEST LABORATORY
MONTHLY ACTIVITIES REPORT

MARCH 1970

DIVISION OF PRODUCTION
AND
HANFORD PLANT ASSISTANCE
PROGRAMS

April 1970

AEC RESEARCH &
DEVELOPMENT REPORT

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PACIFIC NORTHWEST LABORATORY
MONTHLY ACTIVITIES REPORT

MARCH 1970

Division of Production
and
Hanford Plant Assistance
Programs

by the

Staff of Battelle-Northwest
F. W. Albaugh, Director

April 1970

PACIFIC NORTHWEST LABORATORY
RICHLAND, WASHINGTON

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PACIFIC NORTHWEST LABORATORY

MONTHLY ACTIVITIES REPORT

DIVISION OF PRODUCTION AND KANFORD PLANT ASSISTANCE PROGRAMS

W. D. Richmond

ASSISTANCE TO DOUGLAS UNITED NUCLEAR

Summary

Program I

Hybrid simulation of N-Reactor was debugged. Turbine trip transients were made with the system in real-time.

The final report is nearing completion on autoclave exposures of stressed 304 stainless steel specimens and Inconel-steel couples.

Program III

The test assembly to study simulated loss-of-coolant accidents in N-Reactor was fabricated and successfully operated. Present efforts are being directed to provide test pieces with surface roughness and emittance characteristics which approximate N-Reactor fuels.

Process Technology

An anodic hydride removal process in nitric acid and in a nitric acid-fluoride mixture was evaluated by examining the downstream portions of two process tubes removed from KE Reactor after being treated. Hydrogen analyses of the rear five foot portions of the two tubes showed the anodic treatment in nitric acid left a surface layer of dispersed hydride platelets up to 2 mils thick while the subsequent nitric-hydride treatment completely removed this layer and left an etched surface.

A pilot plant test on N-Reactor decontamination using a complexing solution was completed in a test facility loop. Corrosion rates were low, and no localized or preferential attack was noted in the weld specimens.

An analytical study is being performed to develop a method to evaluate the effect of interjunction flow on the thermal hydraulic performance of N-Reactor and SPR fuels. The analysts uses the mathematical model developed in the COBRA-II code to calculate flow, enthalpy and pressure within parallel channels that are coupled by interjunction crossflows. Trial calculations were performed to verify the analytical methods. Sample calculations showed the qualitative effects of interjunction flow on the thermal hydraulic behavior of three parallel channels.

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ASSISTANCE TO DOUGLAS UNITED NUCLEAR

Program 1 - Basic Production

N-Reactor Simulation

(J. D. Lodge, Process Simulation & Analysis Section)

The N-Reactor hybrid simulation was debugged, and turbine trip transients were made with the system in real-time. The entire secondary system resides in the PDP-7 digital computer. The digital section of the simulation has a loop time of 17.4 msec with SIMPL-1 subroutines throughout and little effort to conserve time in this first attempt. The secondary breaks down into six subroutines with the following loop times: steam generator primary side, 5.38 msec; steam generator secondary, 2.38 msec; dump condenser, 1.96 msec; surge tank, 1.05 msec; condensate header system, 3.33 msec; and the steam distribution subroutine and the controller simulator. The controller simulator seems to get less reliable with age, mainly in the IC to compute switching relays.

Corrosion of Inconel Heat Exchanger Tubing

(D. A. Jones, Corrosion Research & Engineering Section)

This investigation was concerned with the stress cracking of 304 stainless steel and possible stress cracking and corrosion of Inconel heat exchanger tubing when exposed to simulated heat exchanger environments. The final correlation of cracking data shows that sensitized 304 stainless steel will crack in the presence of dissolved oxygen and/or dissolved chloride. Cracking was also observed in either alone. Specimens from certain of the operating N-Reactor steam generator tubes showed a somewhat higher frequency of cracking than those from control heats of sensitized 304 tubing. Negligible evidences of cracking or corrosion of Inconel were observed in Inconel-steel crevices. A final report is nearing completion.

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Program III - Environmental and Regulatory Technology

N-Reactor Fuel Temperature Following a Primary System Rupture
(R. E. Collingham and D. E. Fitzsimmons, Thermal Hydraulics Section)

Fabrication of an electrically heated test assembly to study simulated loss-of-coolant accidents in N-Reactor was completed. The entire test apparatus including all systems (electrical, steam, helium, hydrogen collection and data acquisition) was checked out by performing a trial transient run with a dummy test piece. Review of the data indicated that all systems performed satisfactorily.

The autoclaved surface first obtained on the test piece was not considered representative of N-Reactor autoclaved fuel surfaces. This conclusion was based on emittance measurements and visual observations. The surface of the test piece was much rougher than the fuel surface. A sample test piece will be resurfaced to obtain the proper roughness and emittance. Once satisfactory techniques have been developed to adequately approximate the surface conditions of N-Reactor fuels, simulated loss-of-coolant experiments will proceed as funding is available.

Process Technology

Low Temperature Hydriding of Zircaloy
(B. Griggs, Corrosion Research and Engineering Section)

A process for removing surface hydride layers from Zircaloy process tubes by employing an anodic treatment in 3% nitric acid to remove the dense case layer followed by anodic treatment in a 3% nitric acid-1% fluoride mixture to remove the anodic oxide and etch the remaining dispersed hydride platelet layer was tested on several KE Reactor tubes. Two tubes from the test were removed, and the downstream five foot sections were delivered to BNW for evaluation. Tube 2255 had been treated only in 3% nitric acid while 3056 had been additionally treated in the nitric-fluoride mixture. Samples

through the Van Stone flange on both tubes and a ring taken 1 foot upstream on 2255 were examined metallographically. Tube 2255 showed the expected residual layer (<2 mils thick) of dispersed hydride platelets. Tube 3056 showed no such layer and had a clean etched surface except behind the seal near the Van Stone flange, which had not been treated. The case layer and dispersed hydride layer in the untreated area was 19 mils thick, while the surface removal in an adjacent treated area measured 19 mils, which indicated that the surface removal was not excessive. Hydrogen analyses of rings cut from the tube at 1 foot intervals were made by hot vacuum extraction. Two adjacent rings were cut for each position and one of the rings etched 3 to 4 mils per surface in standard nitric-hydrofluoric acid solution. Comparison of the two hydrogen values indicates the uniformity of the hydrogen distribution at that point.

HYDROGEN CONCENTRATION IN ANODICALLY STRIPPED TUBES

Position (Inches from Rear)	Tube 2255KE			Tube 3056KE		
	As Received, ppm	Etched 3 mils, ppm	ΔH , ppm	As Received, ppm	Etched 3 mils, ppm	ΔH , ppm
12	262	126	136	147	142	5
24	279	116	163	119	118	1
36	217	101	116	90	90	0
48	173	92	81	86	83	3
58	132	80	52	61	62	-1

The presence of surface hydride layers in 2255 and the absence of any surface layers in 3056 are clearly shown by the analyses in the above table.

N-Reactor Decontamination

(J. A. Ayres, Corrosion Research and Engineering Section)

A pilot-plant test of a complexing decontaminating solution was completed in TF-9 loop this month. The solution contained diethylenetriaminepentaacetic acid (DTPA), citric

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acid, and hydrazine. The solution at 85°C for 2.5 hr successfully cleaned the sample, an N spacer. Corrosion rates were low, and no localized or preferential attack was noted at the weld specimens. As in the previous test using ethylenediamine-tetraacetic acid (EDTA), water-line staining was noted, but this resulted from the operating procedure and would not occur in an actual reactor decontamination. The loop has no provision for proper dilution and pH adjustment after the decontamination. In a reactor the decontaminant, after recirculation for the desired time, is displaced with a dilute ammonia (or hydrazine) solution to bring the pH to 10 before the system is opened to the air.

In laboratory tests variation between pH 6.0 and 7.75 appeared to have only a minor effect on corrosion rates and decontamination. A pH of 7.75 was chosen as the upper limit since it is the pH at which the citric acid and DTPA are fully neutralized by the hydrazine. The corrosion rate of carbon steel after one hour was about 0.06 mil/hr and then decreased to an average rate of about 0.03 mil/hr after 5 hr. Some variation among the different samples was noted, probably as a result of differences in surface preparation, but is of little practical interest because the rates are so low. Carbon steel samples that had been chemically polished in oxalic-peroxide solutions corroded at an abnormally low rate, as low as 0.01 mil/hr. Coupling with stainless steel did not affect the average corrosion rates of carbon steel, and very little localized attack was discernible at the weld junctions.

Decontamination factors after 2 hr were about 100 except in cases of poor geometry, and after 4 hr, increased to 200. The pH appeared to affect the DF and the rate of decontamination, but no conclusions can be made from these preliminary tests, especially since the experimental conditions are not at all prototypical.

In a loop, or reactor, most of the contaminated film will dissolve off, but a small amount (5%) will be loosened and washed off. This undissolved material will be primarily very fine and will be carried in the solution. At areas of

low flow, some particles may settle, but the amount of settled solids should be low. If this appears to be a problem, a filter or collector (magnetic) should be installed downstream.

Settling of solids may be a problem the first time an extremely dirty reactor is decontaminated, but the activity from these solids should be removed by subsequent treatments, perhaps by a feed and bleed operation during shutdown.

Because of time limitations, the conditions were chosen arbitrarily. It appears that these solutions are safe enough to be used in a reactor and are effective. If the advantages of this type of solution are great enough, in-reactor evaluations should be made. Many questions are still to be answered, however, before the most economical, effective and safe reagent can be specified. If pH is of no great importance, the lower pH (~ 6.0) should be used because less hydrazine is required. Some definite advantage may be gained by replacing part of the hydrazine with a stronger base, such as LiOH. High concentrations may be much more effective, especially for very dirty reactors or for those that have operated a long time since the last decontamination. Some other acid may be used to replace part of the citric acid to improve efficiency, or to decrease cost; a synergistic effect might even be noted. Another complexing agent (ferron, oxine, α -nitroso- β -naphthol, tiron) might be used with the DTPA to increase efficiency. The mixed chelating agents sometimes form extremely stable complexes with certain ions.

Boundary-Value Flow Solution for Fuel Column Thermal-Hydraulic Analysis

(D. S. Rowe, Thermal Hydraulics Section)

The objective of this study is to develop a more complete method for analyzing the thermal-hydraulic performance of the Hanford production reactor fuel columns. The tubular fuel elements used in these reactors direct the coolant through parallel flow paths within a fuel column. Flow communication between the parallel flow channels can only occur through the fuel element junctions. The extent of flow communication

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depends upon the design of these junctions.

Although a quantitative method has not been available to analyze the effect of interjunction flow on fuel column performance, most fuel junction designs allow some interjunction flow within the column. The amount of this interjunction flow and its effect on the thermal-hydraulic performance of the fuel columns is not well known. Analytical methods are needed to help define the importance and role of interjunction flow. This will lead to more accurate predictions of fuel column performance and to more effective fuel junction designs.

An analytical study has been started to develop a method to evaluate the effect of interjunction flow on the predicted fuel column performance. This analysis uses the mathematical model developed for the computer program COBRA-II which calculates flow, enthalpy and pressure within parallel channels that are coupled by crossflows. These crossflows cause a net flow change and transport energy from one channel to another. The model includes various descriptions of crossflow resistance, two-phase flow correlations, friction factors, power splits and channel geometry effects. The boundary conditions required for a boundary-value flow solution include equal exit channel pressure, channel inlet enthalpy and channel inlet flow. For those cases where equal inlet channel pressures are desired, the inlet flows must be adjusted until this condition is achieved. The equations are solved by using finite differences and an iteration procedure whereby the numerical solution sweeps through the channel while holding the upstream and downstream boundary conditions constant.

A computer program has been written to perform the numerical solution. It can accommodate either two or three interconnected channels and, thus, can be used to analyze all production reactor fuel columns. Each channel is assigned a friction factor, flow area, wetted perimeter and a fraction of the total power. The interjunction flow resistance is assigned to each pair of interconnected channels. Nonuniform

axial power distribution can be considered by using tabular values of local/average power versus position along the fuel column.

Several trial solutions were performed for three parallel channels to verify that the basic approach is valid and that the numerical methods are stable. For small values of crossflow resistance, convergence was very fast (1 or 2 iterations) while for large crossflow resistance convergence was slower (20-30 iterations). These calculations showed that boiling was not felt upstream when the crossflow resistance was small; however, when the crossflow resistance was large, boiling caused significant effects upstream. In the latter case sizable pressure differences were created between subchannels at the inlet. To achieve equal inlet pressure for this case requires adjustment of the inlet flow split.

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ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Summary

Program I

Laboratory experiments with the assembly for measurement of nitric acid concentration in the Plutonium Reclamation Facility slag and crucible dissolver have been successful. The next tests will be conducted under simulated process conditions in test facilities at the 324 Bldg.

In the study on Np and Pu monitoring in HAW, analyses of energy resolution and comparisons of isotope and X-ray tube sources were made with the Si(Li) X-ray spectrometer. Collection and separation studies were performed.

In Zirflex tonic studies complexing agents were compared for effect in improving zirconium decontamination in the Purex process. Only ethylenediaminetetraacetic acid (EDTA) and oxalate gave substantial improvement, but both were inferior to fluoride.

Program III

Work continued on the problem of solution convergence in using the Partially Saturated Transient model for simulating leaks in high level waste storage tanks.

The Variable Thickness Transient model was modified to solve steady-state problems in addition to transient problems.

Transducers were placed in a shallow well to measure moisture potential and predict soil moisture movements.

A study was completed to determine the selectivity coefficients for the Ca-Sr ion exchange reaction with a Burbank sand soil.

Eleven of seventeen samples received from the Dow Chemical Company's Rocky Flats Division were given preliminary examinations.

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Continued determination of NO_2 concentrations at three locations near 200 East Area indicate the location of two major sources of NO_2 .

Program IV

Testing of a plant type alpha cell to be used in a study of the variance between laboratory and plant results with cerium activated Vycor alpha sensors showed that the cell was air tight to 20 psi but did show light leakage.

Duolite C-3 which had received a simulated 2-month-use test retained 71% of its capacity for removal of cesium from Redox alkaline supernate.

Preparations were completed for hot cell testing of the proposed zeolite process for purification of B-Plant strontium product. Irradiation studies (with cobalt-60) suggest that radiolysis of complexants may result in a strontium precipitation problem.

A sulfate precipitation scheme for purifying strontium product has shown much promise in laboratory tests and will be evaluated with B-plant product strontium.

An electrodeless conductivity cell for immersed service in Purex plant HAW was constructed and is under test to insure mechanical and electrical acceptability.

Discrepancies in thermal conductivity measurements on waste sludge from ARHCO storage tank samples have been resolved.

Maximum temperatures in air-cooled underground tanks containing radioactive wastes were estimated for 4096 sets of operating conditions.

Strontium-Cesium Waste Packaging Program

- A. Testing continued during the month on the compatibility of cesium and strontium compounds with containment materials.

- B. High pressure (5000 psig) water was ineffective in removing SrF_2 from welded metal surfaces.
- C. Evidence obtained in the determination of the CsCl-KCl-NaCl phase diagram (>80% CsCl) indicate that the solid-solid phase transition may occur at a temperature as low as 311°C .
- D. Additional experimental work on the process for converting $\text{Sr}(\text{NO}_3)_2(\text{aq})$ to SrF_2 continued. An experiment was made to determine the type of filter cake obtained while filtering through a porous cylinder with longitudinal strips that were impermeable to the filtrate. The technique appears suitable for controlling the size of agglomerates of dry SrF_2 .
- E. A calculational model for the effect of cesium loading on potassium decontamination is being developed in a study on cesium purification by ion exchange.
- F. Process equipment for evaporating a CsCl solution to dryness, melting the CsCl , and casting the CsCl into a series of primary containers was operated to determine the corrosion resistance of the Hastelloy C-276 dryer-melter. Corrosion of the Hastelloy C-276 in the area of the vapor-liquid interface has been detectable with ultrasonic probe measurements. Off-gas tracer experiments demonstrated DF's for volatilized CsCl of greater than 13,000 at a filter face velocity of $1\frac{1}{2}$ ft/min.
- G. An experimental test of the efficiency of heat removal from heat sources located in water-cooled storage basins is being conducted.
- H. Equipment for thermal and mechanical testing of prototypical waste capsules is ready for operation.
- I. Estimates of maximum temperatures to be obtained in direct burial of wastes in tunnels were made. A thermal resonance effect due to interaction between adjacent tunnels filled at different times was predicted.

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- J. The prototype impact consolidation loading mechanism for SrF_2 capsules will be operable by the end of March.
- K. In nondestructive testing studies "surface follower" ultrasonic signal gate circuits were developed. Acoustic emission weld monitoring techniques are being evaluated.

Efforts to identify the material in Purex plant uranium product which poisons a platinum catalyst (with respect to uranium reduction) have been unsuccessful.

Study of means of destroying hydrazoic acid without increasing the salt content of process wastes was continued.

Centrifugal contactor runs were continued to test flow-sheets involving use of hydrazine-stabilized U(IV) as a reductant for Pu in the Purex 1BX (1st cycle partitioning) column. Low U(IV) material balances continue to be an unresolved problem.

Evaluation of alternative schemes for solidifying Purex current acid waste has led to the recommendation that the processes for more detailed study be spray and fluidized bed calcination, with the alternative of post-treatment through a melting stage.

Continued review of potential processes for the solidification of the bulk salt (low heat) Purex wastes confirmed the need for assessment of safety aspects (potential for dispersal) of candidate solidified waste products.

Process Technology

A study was initiated to determine the corrosiveness of Purex dissolver solution as a function of the Al/F mole ratio. With a fluoride and Zr(IV) content of 0.1 and 0.03M respectively, the corrosiveness of "initial" dissolver solution was determined to be excessive at Al/F ratios ≤ 1.0 . Corrosion of 304L was slight in "initial" dissolver solution containing 0.2M Zr(IV) and no $\text{Al}(\text{NO}_3)_3$ (Zr/F=2.0).

Huey test results were evaluated as a function of condenser type and boil-up rate. With good quality 304L the corrosion rate was independent of both condenser type (Allihn, Friedrichs, cold-finger) and boil-up rate.

Precipitation characteristics of solutions containing the nitrates of Cs, Na, and K were determined. Successful mixer settler runs were performed to test flowsheet modifications aimed at improved partitioning of uranium from plutonium in the CC Column of the Plutonium Reclamation Facility.

Flocculating agents were evaluated with respect to improving filtration characteristics of ZPPR oxide dissolver solutions.

Experimental work with a lucite model of a section of the Purex annular dissolver was completed. Information was gathered for the use of the Purex dissolver to process Plutonium Recycle Test Reactor type fuels.

Work in support of the Plutonium Reclamation Facility involved design modification of filter socks for removal of undissolved material from the circulating dissolver streams.

Uranium(IV) is being prepared for a Purex Plant test of U(IV) as a plutonium reductant in the 1BX (1st cycle partitioning) column.

Tests of a thermal flow meter for organic service are being conducted. Although nominal sensitivity, repeatability and accuracy are comparable to the conventional plant rotameter, temperature excursion rates can significantly affect accuracy despite temperature compensation.

Detector capabilities for U-Pu isotopic analysis are being investigated.

The ^{14}C content of Hanford fuels was calculated for several typical or expected reactor conditions. Each gram of carbon in the fuel will result in from 0.008 to 0.021 microcuries of ^{14}C , depending on exposure and enrichment.

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A study to measure a series of soil-chemical parameters for a group of Hanford wells was continued.

Pump tests were run on six wells during the month and all indicate that the confined aquifers have a low permeability.

Transmissibility and aquifer bottom maps were redigitized, and the data are being converted to IBM cards for use in simulation models.

The piezometric-sampling pump was field tested and two samples obtained.

Core samples were taken around C Tank Farm. No contamination was observed more than 20 feet below the surface.

The components of a sludge level detector have been assembled and checked, but the scanning of a waste tank has been delayed by waste processing problems at ARHCO. The scanning is tentatively scheduled for early April.

Modifications have been designed, fabricated, and implemented on the ambient air NO_2 filter photometer. Preliminary tests are encouraging.

Further exponential experiments were performed for establishing the effect of fixed neutron poisons (stainless steel Raschig rings containing 1 wt% natural boron) on the criticality of plutonium nitrate solutions. Exponential experiments were performed with 418 g Pu/liter nitrate solution in an 18-in. diameter, water reflected cylindrical vessel containing stainless steel Raschig rings. Previous measurements in this same system with 275 g Pu/liter solution resulted in a large negative buckling ($k_\infty < 1$). A large negative buckling was also obtained for the highly concentrated, 418 g Pu/liter solution, establishing that criticality is not possible for such a system in any geometry.

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Program I - Basic Production

Plutonium Reclamation Facility Dissolver Acidity Sensor
(D. P. Brown, R. C. Weddle and W. G. Spear, Instrument
Research and Development Section)

The use of process stream conductivity and specific gravity measurements to determine the concentration of the nitric acid solutions in the slag and crucible dissolver should achieve the desired concentration measurement accuracy of $\pm 0.5M$.

Previous laboratory calibration and testing on the combined sensor assembly showed capability of measuring the specific gravity from 1.0 to 1.36 and the conductivity from 0.03 to 0.9 mhos/cm over the expected acidity range of 0.1M to 6.0M. Further tests on the specific gravity portion of the assembly evidenced minor data repeatability dependence on flow versus no-flow conditions. This required some process flow for consistent results. Since the sensor will be employed in an in-line mode during dissolver operation, effects on the system indication should be minimal. Based on the successful results to date in the laboratory, the system will be moved as soon as possible to the 324 Building for additional tests under simulated process conditions.

Monitoring of Np and Pu in HAW

(M. C. Lambert, Analytical Chemistry and Instrumentation
Section)

Energy resolution of the Si(Li) X-ray spectrometer was 530 eV at the energy of Pu $L\beta_1$ fluorescence. This is theoretically capable of resolving Np and Pu $L\beta_1$ lines. Some unknown behavior has been observed, however. Resolution of the order of 500 eV has been repeatedly confirmed for isolated X-ray lines, but when a pair of lines are separated by about 800 eV, some line pairs are well resolved and others are not. Resolution decreases as either the X-ray energy or the count rate increase.

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Comparisons were made at different energies between the X-ray intensities obtained with an Am-241 source and an X-ray tube source to guide in the selection of a source to put in the Purex sample gallery.

The powerful collection technique of reverse phase chromatography was rejected because of the undesirability of introducing solids into the Purex sample gallery. Liquid-liquid extraction for collection of Np from extreme dilutions (as in HAW) with subsequent concentration to a level sufficient for direct X-ray fluorescence measurement was extremely difficult. Consequently, the possibility of an automated spectrophotometric measurement was again investigated. Colorimetric determinations of both Np and Pu with Arsenazo III requires 25-fold to 50-fold less concentration than does direct X-ray spectrometry.

Some separations needed for either analytical method are identical. Np and Pu must be separated from each other and from U, Zr, and Mo. Other minor interferences are different for the two methods. A scheme of two or three liquid-liquid extractions is being developed from data obtained with different solvents. Some separations and extractions are more efficient from HCl than from HNO₃ media, and the colored Arsenazo complexes are seriously interfered with by HNO₃. But this presents no serious problem since the Np and Pu can be back-extracted from the first organic phase into any desired media.

Zirflex Tonic Effect

(J. L. Swanson, Applied Chemistry Section)

Earlier work showed that addition of fluoride or oxalate to feeds containing uranium carbide reaction products gave a marked reduction in the amount of unscrubbable Zr present in extracts of such feeds. Also, addition of fluoride to scrub solutions was found to reduce the amount of unscrubbable Zr (this effect was much less pronounced with oxalate). Recent tests of other possible complexants showed ethylenediaminetetraacetic acid (EDTA) to be effective when present in the extraction step but to have little effect when added to the

scrubs. Tartronic acid was slightly effective. Citric acid, barbituric acid, mandelic acid, nitrilotriacetic acid (NTA), hydroxyethylenediaminetriacetic acid (HEDTA), and diethylenetriaminepentaacetic acid (DTPA) had no effect. Phenylarsonic acid gave increased extraction of Zr.

A few experiments were performed to determine how much fluoride is required to give a substantial decrease in the extraction of Zr into 30% TBP-NPH in the absence of uranium. Such data are of interest in devising flowsheets for the recovery of Pu from Np targets. At a HNO_3 concentration of 2.5M, Zr distribution coefficients of 0.13, 0.05, and 0.01 were measured with no fluoride, 0.001M $^\circ\text{F}$, and 0.01M $^\circ\text{F}$, respectively. At 4.2M HNO_3 the respective values were 0.58, 0.15, and 0.02. Niobium distribution coefficients were found to be higher in the presence than in the absence of fluoride but were still quite low (less than 0.05).

Radionuclide Transport Model

(M. H. Karr, Earth Sciences Section)

Work continued on modifying the PST (Partially Saturated Transient) flow model to minimize the dependence of solution convergence on the time step increment. If successful, the modification will result in more efficient solution of simulated high-level waste tank leaks and pipeline ruptures.

Software coding of the portion of the transport model describing the real-time movement and concentration of macro-ions in the Hanford groundwater system was initiated. The macro-ion concentrations along with soil sorptive characteristics will be used to calculate the time-dependent trace-ion sorption isotherm. Progress was excellent, and initial debugging and testing are planned shortly.

Results from the testing of the recently developed weighted average interpolation scheme for converting randomly spaced data to uniformly spaced data were not satisfactory for the type of data encountered in the Radionuclides in Soils Research Program. More effort will have to be expended to develop a satisfactory interpolation scheme.

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The VTT (Variable Thickness Transient) flow model was modified to solve steady-state problems in addition to transient problems. The steady-state option will be used to quickly determine the end effect on the Hanford flow system of changes in waste management practices or environmental changes. The transient option will be used to provide detailed supplemental information (when necessary) on the course of changes with time.

Input data were prepared for the VTT flow model system of irrigating sections of land north of Gable Mountain with warm water from N Reactor. Model verification and solutions will be obtained once the transmissibility and aquifer bottom data have been converted to IBM cards.

A literature review on the diffusion equation was completed. Rough draft copies of the report were distributed for review. This report suggests that irreversible thermodynamics be used to model the drier ranges of the Hanford soils and outlines a research program needed to implement the approach.

Field Measurement Methods

(M. H. Karr, Earth Sciences Section)

Literature was reviewed on existing methods of measuring hydraulic characteristics of soils. From this review of literature rough draft laboratory procedures have been written which will enable laboratory measurement of hydraulic conductivity in the drier range and also some of the osmotic characteristics. Equipment related to this work was ordered. Literature was also scanned to obtain computational methods of characterizing hydraulic conductivity from limited data. These methods will be studied in the coming months.

Instruments (psychrometers and temperature sensors) were successfully placed in a 14 ft 4 in. hand-augered hole, located near Well No. 699-32-49A. It is hoped that these instruments will help give qualitative implications on the depth of rainfall percolation. Plans are underway to make a similar installation on a 320 ft well. Instruments are being prepared for the 320 ft well, and methods are being considered to prevent failures

similar to those previously encountered on deep wells.

Soil-Waste Interrelationships

(R. C. Routson and R. J. Serne, Water and Waste Management Section)

A study was completed to determine the selectivity coefficients (ion exchange constants) for the Ca-Sr ion exchange with a Burbank sand soil. Data were found to fit the general mass action expression

$$\frac{(A)}{(B)} = K_{A-B} \left(\frac{[AX]}{[BX]} \right)^n \quad (1)$$

where

K_{A-B} = selectivity coefficient for the reaction
 $AX + B \rightleftharpoons BX + A,$

n = empirically determined exponent,

AX and BX = concentration of components A and B
sorbed on the solid phase, respectively, and

(A) and (B) = the solution activity of ions A and B,
respectively.

A plot of $\log[(A)/(B)]$ versus $\log[AX/BX]$ was found to be linear. The slope of this plot is equal to the exponent n , and the intercept is equal to the log of the selectivity coefficient. The selectivity coefficient and n were found to be 0.13 and 4.28, respectively.

Atmospheric Release of Radioactive Particles

(L. C. Schwendiman and J. Mishima, Air Pollution Chemistry Section)

Each of thirteen specimen surfaces obtained from Dow Chemical Company's Rocky Flats Division were given a preliminary examination, and a detailed examination procedure was outlined for each specimen. Contamination levels were determined roughly

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with alpha particle survey-type instruments.

Five specimens which could easily be sectioned (paper, plastic films) were examined microscopically.

Gaseous Waste Assistance

(M. A. Wolf, Atmospheric Analysis Section)

Examination of NO₂ concentration data at three sites in the vicinity of the 200 East Area continued. Mean concentrations at each site were determined as a function of wind direction at the 200 ft level of the Hanford Meteorology Tower for all hours and for hours having average wind speeds in excess of 4.5 miles/hr. When the mean concentration wind roses are plotted in the proper geographic position, two major NO₂ sources are indicated as the Purex Plant and a source to the north of Purex.

Program IV - Waste Management

Alpha Scintillating Glass

(O. H. Koski, Analytical Chemistry and Instrumentation Section)

The plant experience with cerium activated vycor sensors is at variance with results observed in the laboratory. To clarify this variance, a study will be conducted through the duplication of plant sensors, cells, stream samples and electronics.

An alpha sensor cell made to duplicate plant construction was pressure tested and characterized with respect to alpha response on three types of typical dry plant alpha sample mounts. Although the cell was air tight to 20 psi, a leakage of light was noted.

Ion Exchange Removal of Cesium from Redox Alkaline Supernatant

(J. R. Lundquist, Applied Chemistry Section)

Further studies were made on the degradation of Duolite C-3 in a 2.7M hydroxyl ion environment. For data simplification

the time of the test was converted to equivalent plant months of operation. A Tank 102 SX synthetic solution (diluted 1 to 1) was passed over the resin, and the 5% cesium breakthrough was determined. The breakthrough occurred at 38 column volumes with new resin, at 34 column volumes after 2 months contact and at 27 column volumes after 8 months contact. Thus the resin retained 71% of its original capacity for cesium after contact with hydroxyl ion for 8 months, which corresponds to 134,000 Ci of ^{137}Cs /3000 gallons of resin.

Purification of Strontium by Ion Exchange
(L. A. Bray, Applied Chemistry Section)

Initial laboratory studies were completed preparatory to hot-cell demonstration of the feasibility of purifying strontium from the B-Plant solvent extraction product by Linde 13-X inorganic exchange. Tracer loading and elution studies were completed using ^{85}Sr , ^{54}Mn , ^{28}Mg , and ^{45}Ca . Decontamination factors for the 0.001M Sr feed were determined after loading with 370 cv (column volumes) and eluting with 25 cv of 3M ammonium acetate. The DF values for Mn, Fe, Pb, Ca, and Mg were 480, > 90, 271, 38, and 20, respectively. Operation of two columns in series (same total column volume) resulted in an increase in DF. The DF's for Mn, Ca, and Mg were 650, 71 and 43 respectively, for the first section (100% breakthrough) and 398, 43 and 20, respectively, for the second section (5% breakthrough).

Studies with ^{60}Co were completed to determine irradiation stability of the complexed LBP feed and the 3M ammonium acetate - 0.015M Sr product. The Sr precipitated from solution as irradiation-produced CO_2 was formed. The precipitate in the feed sample was dark brown and difficult to dissolve in HNO_3 . A white precipitate formed on the vessel walls in the acetate solution and broke off in large sheets. It dissolved readily in acid with CO_2 evolution. The results indicate that the complexed LBP feed is not stable to radiation above 1 to $4 \times 10^6 \text{R}$. The strontium initially found in the 3M acetate was completely precipitated after exposure to $2 \times 10^8 \text{R}$.

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Purification of Strontium by Multiple Sulfate Precipitation
(L. A. Bray, Applied Chemistry Section)

An alternate strontium purification flowsheet was proposed. The B-plant solvent extraction product is diluted to 0.015M Sr (as compared to 0.001M for ion exchange) and precipitated without carrier as the sulfate. Two cycles of precipitation and metathesis are required. A final adjustment to pH 10 (to remove rare earths) is also recommended. Decontamination factors through the two sulfate precipitation steps for Mn, Fe, Ca, Mg, Cd, and Na were > 14,500, > 35, 52, 393, 1500, and 1.8, respectively.

Electrodeless Conductivity
(O. H. Koski, Analytical Chemistry and Instrumentation Section)

The mechanical structure which houses the sensing elements of the immersion type electrodeless conductivity probe to be installed for HAW stream analysis was fabricated. Tests showed the need for careful toroid-to-shell and wire-to-toroid insulation. Also irregularities in the dimensions of ceramic parts resulted in assembly difficulties.

Thermal Conductivity Measurements
(D. G. Bouse, Applied Chemistry Section)

Conflicting results in measurements of the thermal conductivity of a sludge sample from Tank 116 TS were obtained with various sample preparation techniques. The value reported for this material in January 1970 was 0.468 BTU/(hr)(ft²)(°F/ft), measured at 25°C. The sample had been wetted and dried into a hard cake that was noted later to contain 3 salt layers. For a more homogeneous sample, the sludge was again wetted to form a thick paste which was then dried. Thermal conductivity measurements of this material were much lower than the previous sample. Inspection of the probe after removal from this sample revealed large voids under the hard crust, which had separated the probe from the sludge near the center of the sample. At the request of the program sponsor the sludge sample was again wetted (it was necessary to dissolve much of the sample in the

process of removing the probe) and redried. A soft layer about 1/2 in. thick formed on the top of this sample. The lower portion, in which the probe is embedded, appears to be very hard and uniform. Thermal conductivity of this material is $0.453 \text{ BTU}/(\text{hr})(\text{ft}^2)(^\circ\text{F}/\text{ft})$, measured at 29°C . The slight physical differences between the initial and final samples and the fairly close agreement between the thermal conductivity values suggest that the above values are accurate.

Underground Heat Transfer in Waste Tanks
(G. Jansen, Chemical Engineering Section)

Calculations of maximum temperatures in solidified wastes in underground tanks were completed for a wide variety of operating conditions. The FORTRAN V computer program SLUCE was used to estimate maximum temperatures for 4096 combinations of operating conditions with varying air rates, cake thickness, heat generation rates, heat generation distributions, and the effects of water in the system. Among the conditions considered were heat generators beneath bulges in the tank liner. These results may be applicable to the continued surveillance of Tank 105A in the ARHCO waste tank farm.

Comparison of the calculated results with the operating conditions in Tank 108SX, which contains solidified, air-cooled wastes, is underway. Initial considerations in this comparison are that the measured thermal gradients in the cake are not typical of what one would expect from uniformly distributed heat generators, that heat storage in the soil beneath the tank may play an important part in the transient conditions, and that heat losses along the temperature probe may not be negligible.

Cesium-Strontium Waste Packaging Program

The following lettered items present progress on tasks in support of the Cs-Sr waste packaging program.

A. Compatibility Studies

(H. T. Fullam, Applied Chemistry Section)

Testing continued on schedule in all the compatibility

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studies. The six-month radioactive test couples were sectioned, and photomicrographs are being obtained. Visual examination of the test couples indicated no excessive attack on any of the specimens. Examination of the welded Hastelloy C - CsCl and welded 316L - CsCl couples tested for four months at 400°C shows very little evidence of attack. The welded metal surfaces appear to be no more susceptible to attack than unwelded surfaces.

B. Decontamination Studies

(H. T. Fullam, Applied Chemistry Section)

Work continued on decontaminating SrF_2 from welded metal surfaces. No satisfactory decontamination method was found. High pressure (5000 PSI) water spraying was found to be ineffective. A nine-minute spray reduced the SrF_2 contamination to about 2% of that originally present. Additional spraying would not reduce the residual contamination much below the 2% level. Electrochemical and ultrasonic cleaning in 0.1M HNO_3 was not effective in reducing the contamination level of the samples, which were water sprayed, below about 1%. Water spraying at 5000 PSI with a -325 mesh magnesia sand was recommended by the ARHCO Decontamination Facility, and this will be tried if it is decided that such a system can be used in the Packaging Plant.

C. Phase Diagram Studies

(H. T. Fullam, Applied Chemistry Section)

Work continued on phase diagram, phase transition temperature, and thermal expansion studies for the system CsCl-NaCl in the high CsCl range (> 80 wt%). Preliminary evidence indicated that the CsCl phase transition temperature is as low as 311°C when substantial amounts of KCl are present.

D. SrF_2 Process Studies

(L. K. Mudge, Chemical Engineering Section)

A flowsheet for conversion of $\text{Sr}(\text{NO}_3)_2(\text{aq})$ to SrF_2 suitable for encapsulation is being developed. Drying SrF_2 on

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a porous metal filter produces large agglomerates which become extremely hard after sintering. The size of such sintered agglomerates could cause problems in consolidation of radioactive SrF_2 into containers.

A method for controlling the size of dried SrF_2 agglomerates was tested and involved putting longitudinal strips of 1/2 in. tape, which was impermeable to the filtrate, on a porous metal filter. The tape was spaced on 1 1/2 in. centers. With such a filter a SrF_2 filter cake which consisted of strips 1/4 in. thick by 1 1/4 in. wide was obtained. Arrangement of impermeable strips in a checkerboard design on a filter should be a satisfactory method for controlling the maximum size of dried SrF_2 agglomerates, if such control is needed.

E. Cesium Ion Exchange

(G. Jansen, Chemical Engineering Section)

Basic equilibrium and kinetic parameters are being determined for use in calculating ion exchange breakthrough curves in purification of cesium on an inorganic cation exchanger. Equations involving the exchange of cesium, potassium, sodium, and ammonium ions are being programmed for approximate computer calculation of the behavior of these ions during loading, scrubbing, and elution. A bilinear, kinetically limited reversible exchange model is being used to include the effect of high cesium loadings on potassium decontamination factors. The partial ionization of ammonium ion is being included in the model. A trial hand calculation was made including all the elements to be used in the computerized model.

F. CsCl Melt Casting

(L. K. Mudge, Chemical Engineering Section)

A process for casting molten CsCl into a series of primary containers is being developed for use in the projected Hanford Waste Management Plant. Techniques for filling a series of containers were developed, and the equipment is being tested to determine operating parameters and equipment corrosion performance.

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Corrosion of 0.004 in. to 0.007 in. has been measured ultrasonically in the area of the liquid-vapor interface in the Hastelloy C-276 dryer-melter. The dryer-melter was operated at temperatures of 100 to 120°C for about 50 hr for evaporation of a pH 4 solution of CsCl and at melt temperatures of 600 to 700°C for about 10 hr. No corrosion of 304 stainless steel was noted by this technique after about 3 times as much operating time.

Tracer studies with $^{134}\text{CsCl}$ were conducted to test the efficiency of an absolute filter (MSA, Type H, No. 76876, 99.97% efficient for 0.3 micron particles) for removal of volatilized CsCl from the melter off-gas stream. At a filter face velocity of 1 1/2 ft/min, DF/s of > 13,000 and > 22,000 across the filter were obtained in two separate runs. After completion of the two runs the absolute filter was washed, and the amount of CsCl retained by the filter was determined. Comparison of the amount of CsCl on the absolute filter to the amount that passed through it gave a combined DF for the two runs of > 147,000. Handling of the off-gas from the CsCl melter does not appear to be a difficult problem.

G. Storage of Cesium and Strontium

(G. Jansen, Chemical Engineering Section)

One concern in storage of cesium and strontium capsules in a water basin is the possible occurrence of dead spots in water circulation that might allow the container to overheat. A possible solution to this problem is the introduction of cooling water near the floor of the basin. The main stream of water then passes horizontally across the capsule array located near the bottom of the basin and then exits near the floor of the basin. Flow would be directly across the capsules and a large recirculating eddy would be created in the upper part of the basin. The location of the dead spot would then always be predictable and would not be near the capsules. The occurrence of local dead spots downstream from each capsule might cause local overheating. An experimental measurement of the local temperature variations during these operating conditions is underway. A 1 KW heater with capsule dimensions is placed in a transparent pool. Water is passed over the capsule at 25 gpm/ft of pool width. The local temperatures near

the capsule and the circulation patterns are observed. At the present time, no temperature measurements have been made in the system.

H. Capsule Integrity Testing

(J. Dunn, Chemical Engineering Section)

Thermal and physical tests of the double walled metal capsules containing cesium chloride and strontium fluoride are necessary to demonstrate capsule integrity.

All of the stainless steel tubing for capsule fabrication was forwarded to J. Wormeli of ARHCO for fabrication. All equipment for capsule testing was assembled, and experimentation will begin in April.

I. Direct Burial of Packaged Wastes

(G. Jansen and L. G. Niccoli, Chemical Engineering Section)

Heat transfer calculations were made with FORTRAN V computer program DIBUW to determine the maximum transient temperatures in underground tunnels containing direct-buried wastes. In the calculations, a tunnel was simulated by a long line source with a heat generation rate of 280 Btu/(hr) (ft of tunnel length), corresponding to the placement of 1 kW heat sources at 10 ft intervals along the tunnels. Single decaying isotopes with half lives of 30 years were assumed in the calculations.

The most important variables affecting the maximum temperature are the heat generation rate per unit length of tunnel, the spacing and number of adjacent tunnels, and the thermal conductivity of the surrounding medium. A tunnel diameter in the range 2 to 4 ft and a depth below grade in the range 60 to 100 ft had only secondary effects on the maximum temperatures.

An unexpected "thermal response" effect dependent on the schedule for tunnel filling is described in the following manner. Filling all the tunnels at once does not result in the worst thermal conditions. For example, if two tunnels with a 4 ft diameter are placed 25 ft apart in soil with a thermal conductivity of 0.35 Btu/(hr) (ft²) (°F/ft), a temperature

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wave proceeds from each source. If the tunnels are filled with a 30 year half life material at an initial heat generation rate of 280 Btu/(hr)(ft of length) at the same instant, the temperature at the wall of one tunnel on a line between their centers will receive two waves of temperature rise. The first wave to reach the wall is from the closest tunnel and peaks at 356°F and 10.5 yr. The second wave from the other tunnel peaks at 119°F and 20.5 yr. The maximum temperature, which is the sum of the two waves, does not occur at the time of either individual maximum temperature and reaches a value of 470°F at 13 years.

If the far tunnel is filled 10 years before the tunnel whose wall is being considered, the peaks of the two temperature waves arrive simultaneously at this wall, and a maximum temperature rise of 485°F is reached 10.5 yr after the closest tunnel is filled and 20.5 yr after the far tunnel is filled. Thermal waves from neighboring tunnels on both sides could reinforce this effect by adding their own contributions if they were at appropriate spacings and filling times.

The importance of the effect was not investigated for practical spacings and fill intervals; however, the condition should be avoided if possible. It seems reasonable to expect that there exists an optimum filling schedule and spacing that may possibly include nonuniform spacing and unequal filling intervals, or at least there must be a schedule that does not fill adjacent tunnels at the wrong intervals.

J. Strontium Fluoride Densification

(K. R. Sump, Materials Development Section)

The machining and sheetmetal work on the prototype loading mechanism for compaction of SrF_2 was completed. The installation work is presently in progress and should be completed in March. To date, no serious faults have been observed in the loading mechanism.

Two Al_2O_3 rectangular trays for sintering SrF_2 were received, and eight more are expected in a few weeks. These

trays are available from stock; however, at the present time the vendor is having difficulties in fabrication. The two trays received had distortions and were oversize to the extent that they would not fit the holder in the loading mechanism.

Thermal shock tests on the trays were conducted by putting the trays directly in a 400°C furnace, obtaining uniform tray temperatures, and air cooling upon removal. This process was repeated at increased temperature increment of 100°C. One boat was loaded with Al_2O_3 , and the other contained 800 g of SrF_2 . Both boats survived up to and including the 800°C test and failed at 900°C.

K. Nondestructive Testing Studies

(R. W. Steffens, NDT Systems Engineering Section)

NDT support of the ARHCO strontium and cesium waste encapsulation program continued with the development of "surface follower" ultrasonic signal gate circuits and the initiation of acoustic emission weld monitoring technique evaluation.

The "surface follower" ultrasonic defect signal gate is a modification of a standard gate which permits referencing the gate position to the ultrasonic reflection from the outer surface of the test piece. When the defect gate is positioned at a constant time after the surface reflection, the transducer test piece water path distance can vary without disturbing the ability to maintain the test parameters. Ability to perform the ultrasonic test with water path variances reduces the stability requirements of the remote mechanical scanning equipment and thus simplifies the remote test apparatus.

In situ weld monitoring techniques are being evaluated in a cooperative effort between ARHCO weld personnel and BNW NDT staff. Welds fabricated with the automatic program ARHCO gas-tungsten-arc weld station have been monitored, and the ability to detect extensive catalyst induced cracking was established.

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Alternative Purex Reductant-Platinum Catalyzed Hydrazine
(J. L. Swanson, Applied Chemistry Section)

Identification of the substance that poisons the Pt-catalyzed reduction of U(VI) to U(IV) and is present in the sample of Purex uranium product solution was not accomplished. The poison remained in the supernate after crystallization of about half of the uranyl nitrate hexahydrate (UNH) from a portion of the sample. Spark source mass spectrographic analysis of the two fractions (crystal and supernate) failed to show any significant differences in impurity content.

The poison was not removed by passage through a bed of activated alumina or carbon and was not destroyed by digestion for several hours in about 5M UNH at the solution boiling point. Recycle of effluent back through the catalyst bed (after catalyst regeneration) showed that the poison was removed effectively by the bed.

Hydrazine Reaction Products
(J. L. Swanson, Applied Chemistry Section)

The study of means of destroying hydrazoic acid without increasing the salt content of the waste continued. No reaction was observed between azide and permanganate in carbonate solution, though reaction was observed in acid solution. Results of azide-peroxide and azide-sulfite reaction studies are not clear-cut.

Hydrazoic acid decomposed (presumably to ammonia and nitrogen) fairly rapidly when an acid solution was contacted at 50°C with platinum catalyst pellets. The reaction did not go well in carbonate solution. Whether the reaction will occur with hydrazoic acid in TBP-NPH is presently not known.

Alternative Purex Reductant - U(IV)
(D. G. Bouse, Applied Chemistry Section)

Centrifugal contactor runs were continued to test flow-sheets proposed to demonstrate the use of hydrazine-stabilized uranium(IV) as a reductant for plutonium in the Purex 1BX (1st cycle partition) column.

Discussions held with the program sponsors suggested three additional centrifugal contactor runs to be made prior to the actual Purex plant demonstration run, presently scheduled for late this month. The first run, to determine if Fe(II) is oxidized in the centrifugal contactors as the U(IV) appears to be, was completed. Analyses from this run are incomplete; however, indications are that <0.1% of the Fe (II) introduced in the LBX stream reports to the aqueous product as Fe(II). Partitioning of plutonium and uranium appears to be satisfactory.

Other runs to be completed by March 20 include: a) a run with U(IV) in which the hydrazine concentration of the end streams is determined and b) a run with increased U(IV) addition.

Solidification of Current Acid Wastes

(J. E. Mendel, Engineering Demonstration Section)

The tabulated data assembled last month on the candidate solidification processes was used to make: (1) a qualitative comparison of estimated capital and operating costs; and (2) a forced decision analysis. The forced decision analysis considered costs but also included an evaluation of several other factors less susceptible to quantification, such as process flexibility, operating safety, product quality, and required effluent treatment. Two processes, fluid bed and spray calcination, were shown to have clear economic and technical advantages over the other reviewed processes, which were pot calcination, rising level glass, phosphate glass, FINAL (the English rising level glass process), and pot vitrification (the French batch process).

The outstanding advantages of the two calcination processes result because (1) they are continuous and thus scaled up easily and (2) because they are flexible, both as to the range of waste constituents which can be processed and as to the types of product which can be produced. Potentially volatile waste constituents, such as sulfate and fluoride, can be handled because the processes operate at relatively low temperatures.

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The calcines from both processes can be given post-treatments easily to make a less dispersible final product form. In-pot melting, using a glass frit flux, appears especially suitable for this purpose. Dispersion of the calcine in a metal also merits consideration.

Solidification of Bulk Salt Waste

(W. K. Winegardner, Engineering Demonstration Section)

Additional review of potential processes for the solidification of the bulk salt (low heat) Purex wastes confirmed the need for careful assessment of the safety aspects (potential for dispersal) of candidate products. Incorporation of radioactivity into asphalt is the only process with plant experience which results in a product that is relatively inert to water. Possible techniques to remedy the problem of oxidizing salts in asphalt include denitration, conversion to sulfates, and the use of plastic instead of asphalt. In addition, the possibility for dispersal by either water or high temperature of any product arising from the incorporation of modified (unmodified) low-heat Purex wastes in asphalt (plastics) should be evaluated.

Evaporation-crystallization and either fluidized bed or pot calcination processes could be adapted to solidify the bulk salt waste stream. The products, however, could be easily dispersed by water if the containers were breached. Conversion of either salt cake or calcine into glass is feasible but would require some additional process development.

The tendency for fluoride, which is present in the reference bulk salt waste, to volatilize in the high temperature glass-forming processes must be defined before those processes can be considered further.

Another possible technique, if a high quality solid is desired, is incorporating the fluidized bed product into an insoluble matrix. Again, additional R&D studies would be required.

If processing conditions at the Purex plant can be modified to reduce the salt content and eliminate the fluoride in the bulk salt waste, the possibility of processing the low heat waste stream in conjunction with the high-heat waste stream warrants consideration.

Process Technology

Corrosion Studies

(R. F. Maness, Corrosion Research and Engineering Section)

Data previously obtained by this laboratory indicated the corrosiveness of Purex ZWW solution (aqueous waste resulting from processing Zr-clad fuels) is reduced substantially (> three-fold) by reducing the nitrate content of the solution from about 8.0 to 6.5M. It appeared likely that a reduction in nitrate content could be obtained via a reduced aluminum nitrate addition to the dissolver solution. Thus, 304L corrosion rates are being obtained in simulated "initial", "mid-point" and "final" Purex dissolver solution as a function of $\text{Al}(\text{NO}_3)_3$ concentration with an assumed fluoride and Zr(IV) content of 0.1M and 0.03M, respectively. An additional series of evaluations are being made with fluoride and Zr(IV) concentrations of 0.05 and 0.015M, respectively. Data have been obtained to date only in "initial" dissolver solution ($\text{HNO}_3 = 10\text{M}$). In the solution containing 0.1M fluoride and 0.03M Zr(IV), 304L specimens exposed to the liquid phase as well as specimens exposed to the vapor phase corroded at rates of about 100, 10 and 3 mils/mo at Al/F mole ratios of 0.0, 1.0 and 3.0, respectively. The Al/F mole ratio in current Purex ZWW solution is about 5.5. The corrosion rate of 304L in "initial" dissolver solution was low (about one mil/mo) when the Zr(IV) content was increased from 0.1 to 0.2M (no $\text{Al}(\text{NO}_3)_3$ present).

Huey test results were previously evaluated as a function of condenser type and solution boil-up rate. Accelerated corrosion of poor quality (Huey rate = 2.1 mils/mo) 304L was obtained when an open condenser (vs cold-finger) was used and also when a high boil-up rate was employed.

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The tests were repeated with good quality (Huey rate = 0.7 mil/mo) 304L specimens. The corrosion rate was found to be independent of condenser type (Allihn, Friedrichs and cold-finger) and of boil-up rate.

Cesium Nitrate System Solubilities

(L. A. Bray, Applied Chemistry Section)

The solubility characteristics of nitrate solutions comprised of 0.75M Cs^+ plus varying amounts of K^+ and Na^+ were determined. The system is quite complex, and the results, although reproducible, are not readily systemized.

Plutonium-Uranium Partitioning

(D. G. Bouse, Applied Chemistry Section)

Mixer-settler runs were resumed to test flowsheet modifications aimed at improved CC column partitioning of uranium from plutonium in the Plutonium Reclamation Facility. Previous runs were reported before December 1969. The purpose of the first run in the present series was to demonstrate again the coextraction of plutonium and uranium from "fast" ZPPR scrap dissolver solution and to produce the feed for additional CC column studies. The free aluminum nitrate concentration in the feed for this run was 0.7M, compared with 0.6M in the earlier runs. As a result, the plutonium waste losses were reduced from 0.33 to 0.03%. Uranium analyses are not complete.

Previous CC column studies have shown that an increased plutonium-uranium separation factor can be obtained if the plutonium(III) concentration at the top of the column is increased. This was achieved in previous runs by introducing the CA column product at the top of the CC column, operating with a low acid system (not desirable in actual plant operation) and adding sufficient hydroxylamine nitrate to maintain the plutonium(III) medium. The self-salting effect of high uranium concentration in earlier runs may have contributed to improved plutonium-uranium separation. To test the flowsheet for the top of the CC column under all uranium conditions, a series of CC column runs were made with uranium to plutonium ratios of

2.5, 1.25, and 0.4. Analytical results from these runs are not complete.

Reprocessing ZPPR Scrap

(G. F. Schiefelbein, Applied Chemistry Section)

Difficulty in filtering ZPPR oxide dissolver solution containing MoO_3 solids was discussed previously. The small particle size of the MoO_3 makes direct filtration difficult; filtration rates of about 2.8 ml/min/in² under vacuum are obtained.

Several flocculating agents have been evaluated for the purpose of increasing the filtration rate of ZPPR dissolver solution. Ordinary gelatin out-performed all other agents tested by increasing the rate to 6.3 ml/min/in² when 100 ppm were added to the solution. Precoating the filter cloth with ash-free filter pulp increased the filtration rate to about 5.0 ml/min/in².

Purex Dissolution of PRTR Type Fuels

(W. J. Nicholson, Chemical Engineering Section)

The experimental work to develop information for a criticality evaluation of the proposed dissolution of Plutonium Recycle Test Reactor (PRTR) type fuels in the Purex annular dissolvers and to evaluate techniques of solution removal from the dissolver without particle removal is complete. An oral presentation on the work was made to ARHCO personnel. The rough draft of the final report is being prepared.

Plutonium Reclamation Facility Filters

(J. Dunn, Chemical Engineering Section)

Filtration and recovery of undissolved material from the circulating dissolvers is necessary to reduce waste losses. The successful demonstration of a polypropylene sock filter designed and tested by Chemical Engineering Section personnel has led to installation of two of these units in the production plant. Four more similar filters, which can be more readily disassembled and are of smaller size, are being made for the Plutonium Reclamation Facility.

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U(IV) Preparation for Purex Plant Test

(G. L. Richardson, Chemical Engineering Section)

A Purex plant test of the use of uranium(IV) nitrate as a plutonium reductant in the 1BX (1st cycle partitioning) column has been scheduled for late March. Three batches of uranium(IV) reductant, each containing about 640 lbs of uranium total, were prepared in the 324 Building pilot plant facility for this test by an aluminum powder reduction technique. The first batch contained hydrazine as a holding reductant to prevent reoxidation of U(IV) by nitrous acid generated by aluminum-nitric acid side reactions. About 80% conversion was obtained in this test; however, the product contained an objectionable amount of volatile hydrazoic acid (0.02M) and was rejected for use in the plant because of the physiological hazard of using it in the temporary feed station. The second and third batches were prepared using sulfamic acid in place of hydrazine as holding reductant. The reaction was considerably slower (about 6 hours to completion versus 3 hours for hydrazine) and only 60% conversion was obtained. An additional 1600 lbs of uranium has been obtained for conversion to U(IV) to provide sufficient U(IV) for the plant test.

Organic Flow Meter Tests

(O. H. Koski, Analytical Chemistry and Instrumentation Section)

Test and evaluation of a thermal flow meter in the nominal 50 gal/min range is being conducted in the 324 Building with a view toward prospective use for organic metering service in plant where characteristics such as lack of flow restrictions and packing glands, low pressure drop, and weldable design are desirable. The evaluation is by comparison with two other independent flow metering systems. One system is a conventional plant transmitting rotameter using a solid state, variable reluctance, signal conversion. The other system is a turbine type flow meter. With the flow in series through all the meters simultaneous values are compared. In general the rotameter and turbine meter show a comparative accuracy of 1% from 5 to 60 gal/min.

The thermal flow meter is to be calibrated for spans of 40 to 60 gal/min. Interaction of the thermal flow meter controls (span, gain, temperature compensation and bias) requires extensive testing to validate control settings. In addition a study of the effects of temperature changes from 75 to 150°F demonstrated that the temperature compensation required is a function of the rate of temperature change as well as the temperature and the direction of the change. Heating rate was shown to have a larger effect than cooling rate. Temperature compensation settings are, however, chosen to negate temperature effects without regard to rate of temperature change.

Flow rate errors on the order of 7% of flow value were observed for temperature rate changes of +0.3°F/min with a superimposed warm-up error. Warm-up requires 2 hours with an attendant error of from five to ten percent of the flow value.

U-Pu Isotopic Analysis

(U. L. Upson, Analytical Chemistry and Instrumentation Section)

Since conventional detectors (BF₃ tubes ion chambers, etc.) appear marginal, at best, for U-Pu isotopic analysis, new sensors and state-of-the-art developments are being investigated. Of particular difficulty is the high and variable gamma background, which would tend to obscure the neutron response for any reasonably sized neutron source.

¹⁴C Content of Irradiated Fuels

(H. H. Van Tuyl, Process Technology Section)

Although ¹⁴C is not produced in fission, it is produced by neutron activation of carbon impurities in reactor fuels. The amount of ¹⁴C produced is important because of the increasingly restrictive limits on disposal of radioactive nuclides to the environment. Both of the naturally occurring isotopes of carbon have very low neutron absorption cross sections: 0.0034 barns for ¹²C and 0.0009 barns for ¹³C. The cross section of ¹⁴C is even lower, and its half-life is long (5,730 years). Thus, there is only a slow change in the amount of

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each isotope due to burnup or decay. Formation of ^{13}C from ^{12}C is small compared to the amount of ^{13}C initially present. Thus, the amount of ^{14}C formed is directly proportional to the thermal fluence (integrated thermal flux) and also to the amount of carbon present in the fuel. The equations for calculating the amount of ^{14}C are shown below as series expansions of the more familiar exponential equations. The subscripts refer to the mass number of the carbon isotope, and N^0 denotes the amount initially present.

$$N_{14} = \lambda_{13} t N_{13}^0 \left[1 - \frac{1}{2} (\lambda_{13} + \lambda_{14}) t + \frac{1}{6} (\lambda_{13}^2 + \lambda_{13} \lambda_{14} + \lambda_{14}^2) t^2 - \dots \right]$$

$$N_{14} = \lambda_{12} \lambda_{13} t^2 N_{12}^0 \left[\frac{1}{2} - \frac{1}{6} (\lambda_{12} + \lambda_{13} + \lambda_{14}) t + \dots \right]$$

The transmutation constant, λ , is the sum of the radioactive decay constant and the product of flux and cross section.

The typical reactor conditions used for the Waste Management Technical Manual (ISO 100) were also used for this study. Values were calculated as the ^{14}C content in microcuries per gram of carbon and in microcuries from one ton of fuel containing one ppm of carbon. Checks were made with Program ISOGEN to assure accuracy. The results, listed in the following table, show that the ^{14}C content is essentially directly proportional to the exposure for a given enrichment. Some deviation occurs as the reactor type is changed since exposure is dependent on both thermal and resonance neutron fluences, while ^{14}C production is dependent only on the thermal fluence. As enrichment increases, the amount of ^{14}C formed is decreased for the same exposure. This results from the lower fluence required for obtaining the same exposure with more highly enriched fuel. As a rather good first approximation, the ^{14}C content in microcuries per gram of carbon is

$$^{14}\text{C} = 9.6 \times 10^{-6} \times \text{exposure (MWd/t)/enrichment (\%)}$$

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Calculated ^{14}C Content

<u>Enrichment, %</u>	<u>Exposure, MWd/t</u>	<u>Fluence, n/cm²</u>	<u>^{14}C Content</u>	
			<u>$\mu\text{Ci/g}$</u>	<u>$\mu\text{Ci/ppm}^*$</u>
Natural	640	1.73×10^{20}	0.0083	0.0075
Natural	750	2.01×10^{20}	0.0097	0.0088
Natural	900	2.39×10^{20}	0.0115	0.0104
0.947	900	1.92×10^{20}	0.0092	0.0083
0.947	1000	2.12×10^{20}	0.0102	0.0092
0.947	1200	2.53×10^{20}	0.0122	0.0110
0.947	2200	4.33×10^{20}	0.0208	0.0188
1.25	2200	3.42×10^{20}	0.0164	0.0149
2.1	2100	2.03×10^{20}	0.0097	0.0088

* Microcuries per ton of uranium resulting from one ppm carbon in uranium.

Soil-Chemical Parameters

(R. C. Routson, Water and Waste Management Section)

A study to measure a series of soil chemical parameters for a group of Hanford wells was continued. Parameters which will be measured include: 1) moisture content-moisture potential relationships at 15, 1.5, and 0.33 bars; 2) pH in a saturated paste and 0.01M CaCl_2 ; 3) cation exchange capacity (CEC); 4) mechanical analysis; 5) CaCO_3 content; 6) field moisture content; and 7) porosity. Data on CaCO_3 content with depth for well #1 were reported.

The Confined Aquifer (Surface)

(M. H. Karr, Earth Sciences Section)

Certain wells that penetrate into the upper part of the Beverly Member were test pumped during March, with the results tabulated below:

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Well No.	Pump Rate, gpm	Spec. Capacity, gpm/ft drawdown	Transmissibility, gpd/ft (unit gradient)	Permeability, gpd/ft ² (unit gradient)
699-S11-E12	25	less than 1	in process of calculation	
699-10-E12	50	2	1500	in process
699-14-E6	15	7	5000	80
699-20-E5	15	4	3200	80
699-20-E12	17	3	2600	60
699-24-1	8	less than 1	650	7

The aquifer is clearly of low permeability and transmissibility on the southeast side of the Hanford Project toward which the piezometric gradient indicates the water is moving. The specific capacity data, compared with those from wells in the Columbia Basin Irrigation Project, indicate a decreasing capacity in that direction. The combined data tend to confirm that the wells tested lie along what was once the edge of a major stream (the Columbia River) that turned southwestward to enter the lower Yakima Valley. Only small volumes of water evidently can move southeastward in the aquifer in the down gradient direction.

Data were obtained by single well pumping tests, single well recovery tests, and by drawdown and recovery tests in observation wells, where appropriate. The consistency of conclusions validates the techniques used.

Calculation of Transmissibility
(M. H. Karr, Earth Sciences Section)

Due to the inadequacy of the weighted average interpolation scheme, it was not possible to use the Hanford transmissibility and aquifer bottom data that were digitized at the University of Washington Computer Center. However, manufacturer modifications of the graphical digitizer were successful and the digitizer is now operational. The transmissibility and aquifer bottom maps were redigitized and the previously developed straight line interpolation scheme was used to convert the randomly spaced data to regularly spaced data. The data are presently being converted in IBM cards for use in the models simulating the saturated portion of the Hanford groundwater system.

Field Measurements

(M. H. Karr, Earth Sciences Section)

The Widco Logging Unit was mounted on a new truck during the reporting period, and the portable generator is being overhauled. The equipment should be operational for field use by mid-April.

The hydroseismic system was moved to Well E23-2. Output data indicate that either well conditions, low transmissibility of the aquifer or communication with the uncontinued groundwater surface make this well unsuitable for effective hydroseismic monitoring of earthquakes. Present plans are to install the system on Well E33-11.

A design was formulated for obtaining a continuous core at the 116TX Waste Tank. A shallow pilot hole was scheduled for the first part of April to test procedures.

Seven continuous drive core sample holes were drilled around the C Tank Farm. (Radiation levels of the sample material ranged from below background near the surface to as high as 100 rads at depths below the surface and within a radius of 25 ft from the leak source. No evidence of field detectable contamination could be found below 20 ft from the surface. The presence of a caliche layer was found in all the core holes at about 16 to 18 ft below the surface.) Additional core holes are scheduled to be drilled until the geometry of the leak is effectively defined.

Sludge Level Detector

(A. L. Jackson, Analytical Chemistry and Instrumentation Section)

The sludge level detector system was taken to B Plant for pre-scan testing, and the system performed as expected. The system is ready to scan the settling tank in the AR-vault as soon as operations will permit, tentatively early April.

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Ambient Air NO₂ Monitor

(A. L. Jackson, Analytical Chemistry and Instrumentation Section)

Preliminary modifications to the ambient air NO₂ filter photometer had indicated that a read-standardize optical system would improve the sensitivity and stability of the system. Refined modifications were required before additional tests could be completed.

Several methods of modification were evaluated, and a rugged, economical design was selected. Preliminary tests were favorable, and further tests are being conducted to evaluate critically the effect of these changes upon the development objective.

The Effect of Fixed and Soluble Poisons on the Criticality of Solutions*

(R. C. Lloyd, Critical Mass Physics Section)

Further exponential experiments were performed for establishing the effect of fixed neutron poisons (boron-containing steel and glass Raschig rings) on the criticality of plutonium nitrate solutions. Previously, experiments were performed in 12-in. and 18-in. diameter cylinders with plutonium nitrate solutions containing 275 g Pu/l and stainless steel Raschig rings. For the recently completed experiments the solution was concentrated to about 418 g Pu/l and exponential measurements were made in the 18-in. diameter cylinder containing stainless steel Raschig rings. In all of the experiments the rings contained one wt% natural boron, had a wall thickness of 1/16 in., were 1/2 in. in both length and diameter, and displaced about 27% of the solution by volume.

Vertical traverses through the center of the water-reflected 18-in. diameter cylinder were made with the neutron source located below the cylinder. A preliminary analysis of the data indicated the buckling to be about a $-6000 \times 10^{-6} \text{ cm}^{-2}$ as compared to the $-11,000 \times 10^{-6} \text{ cm}^{-2}$ obtained for the 275 g Pu/l solution in the same system. For the Raschig-ring-free

Also supported in part by Division of Reactor Development under Applied and Reactor Physics.

system the material buckling is essentially constant over the 275-418 g Pu/l concentration range, everything else being constant. However, the higher buckling for the more concentrated solution is to be expected in the Raschig ring systems because of the increase in the neutron energy spectrum. Boron, being basically a thermal neutron poison, becomes less effective as the energy spectrum increases. This results in fewer neutron absorptions in the Raschig rings and consequently a higher buckling.

Although the experiments were performed in cylindrical geometry, criticality would not be possible in any geometry, since the poisoning effect of the Raschig rings was sufficient to reduce the material buckling to less than zero for even the highly concentrated solution of 418 g Pu/l.

The 18-in. diameter cylinder is currently being replaced with a 24-in. diameter cylinder. This larger diameter vessel will permit critical experiments to be performed over a range of plutonium and boron concentrations. Criticality measurements will be made with the 418 g Pu/l plutonium solution after checkout of the system and filling of the experimental vessel with glass Raschig rings containing 1/2 wt% boron. The solution displacement caused by the rings will be 19.3% by volume.

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TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Summary

Columbia River Studies

An extensive series of dye tests at 100 K was continued for the purpose of developing a highly accurate plume mixing model. Arrangements for publishing previously classified data were made. Preliminary design of a dissolved nitrogen model is underway.

Waterfowl Census

Consultations were held concerning waterfowl contamination problems at the 100-K and 100-N waste water areas. Preparation was made for the 1970 Canada Goose Nesting Survey.

Thermal Effects Studies

Predation tests continued. Juvenile S. gairdnerii, acclimated to 15°C, were exposed to a water temperature of 23°C for 12, 25, and 50 minutes. Only the 50-minute fish displayed an obvious loss of escape performance.

Surveillance of gas saturation levels in the Columbia River was initiated this month. Several successful attempts were made to monitor gas emboli via Doppler flowmeter.

Step-wise regression analysis of some of the environmental variables and estimated redd count over the period 1947-69 was completed.

The incidence of columnaris and furunculosis disease at the White Bluffs site is probably at a minimal seasonal level. The higher percentage of fish with columnaris antibodies and titer magnitude suggest a higher incidence of infection occurred at the Wenatchee site than at the White Bluffs site during the summer of 1969. Preliminary sampling

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at a smelting plant site suggests that fish residing in warm water areas of the river may retain antibodies against columnaris and furunculosis for longer periods than in cooler water.

Mortalities in juvenile steelhead exposed since early July to approximate Columbia River temperatures and added increments of 3, 4 and 5°F range from 4.0% in the 5°F increment to 15.3% in the 3°F increment. Cold seasonal temperatures favor greater growth in the warmer lots.

Tests on temperature tolerance of aquatic insects continues. LD50 tests over the range +2.5 to +10°C above ambient give insignificant mortalities.

Dosimetry Technology

The manual TLD reader was cleaned and updated with a new heater and phototube before final transfer on March 30.

A temperature of 150°F gave promising results as a quick anneal to be used instead of reader anneal and a one-month holding period. A dosimeter readout 308 days after exposure showed only 3% decay when compared with a dosimeter readout six days after exposure.

Delivery of the prototype multipurpose dosimeters will be delayed four to five weeks due to difficulties in obtaining the specified plastic.

Review of the previously recommended (January 1969) factors for calculation of the doses to the adult bone marrow and endosteal tissue from ^{32}P incorporated into the bone mineral indicated that the factors were somewhat conservative. Since the factors do not include the small dose contribution from ^{32}P in marrow or blood, no reduction in the dose calculation factors was recommended at this time.

Radiation Protection

Capabilities were developed for in vivo counting of ^{147}Pm in lung and bone. Use of the basic thermo-

luminescent dosimeter was implemented for approximately 4,000 Hanford employees.

Environmental Evaluations

A duck containing slightly more ^{32}P than the one obtained on December 29, 1969, at 100-K trench was obtained at 100-N trench on March 6, 1970. The special waterfowl sampling was extended on March 13 to the 200 and 300 Areas, yielding 56 waterfowl from those areas through March 29. External contamination has been detected on waterfowl taken from four 200 Area swamps since the recent release of strontium to B Swamp. A goldfish was obtained from Gable Swamp on March 27. Analysis of the Columbia River above 100 F has been delayed due to the intensive waterfowl sampling effort. Airborne radioactivity from fallout increased slightly and was about the same level as a year ago.

In general, environmental radioactivity measurements indicated no unusual sources of exposure to the population or environs.

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TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Columbia River Studies

(R. F. Foster, Environmental and Life Sciences Division)

Effects of Reactor Effluent on the Quality of Columbia River Water

(R. T. Jaske, Water Resources Systems Section)

Additional dye releases were made in March at 100 K to test new methods for modeling of prompt mixing in the near vicinity of the release point. This series of tests is designed to quantify the effect of jet buoyancy on the initial mixing characteristics of the plume. Special conditions were arranged with 100 K operations through DUN in order to obtain a series of runs at full reactor flows but at zero power level. All sampling of the dye concentrations was done using remote imagery from aircraft. Good resolution of all data was obtained, and the resulting computer output produced real time concentration gradients in the dye patches as they moved downstream. Runs with hot effluent were planned for April.

The sorting and classifying of data and information collected during this program since 1964 was started in order to publish a collection of information which can be released to the public as a result of new classification rules published in January.

Preliminary planning for the development of the dissolved nitrogen gradient model is under way. Model compartmentation and theory are well developed, and computer programming is being outlined.

Mechanisms of Environmental Exposure

(J. F. Honstead, Radiological Physics Section)

The finished draft of a questionnaire for obtaining seafood consumption data from residents of a coastal community was submitted to the printer. The completed questionnaires include photographs of seafood servings to help obtain better

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estimates of consumption levels. The forms will be used in a survey of communities on Tillamook Bay, Oregon, but are designed to permit their use in other seafood production areas. The possibility of extending the survey to a suitable location on the Washington Coast in cooperation with the Washington State Department of Health and Department of Fisheries is being investigated. An effort is being made to measure the necessary calibration factors for the mobile whole-body counter to permit analysis of samples of seafood by gamma scan. This would permit on-the-spot supplementary sampling of important diet components during the survey in a coastal community.

Available means of measuring environmental dose were studied to permit the collection of this type of information during surveys along the Oregon coast. The instruments studied included pencil type ion chambers, calcium fluoride (Dy) TLD chips, and a 2 in. x 2 in. sodium iodide crystal on a 100-ft cable. A preliminary examination of pencils was made to screen out those giving erratic results or having a high background. A test of three sets of pencils charged to 20, 60 and 100 volts and exposed to background radiation levels for 10 days permitted an estimate of the leakage that could be expected. The results indicated an average leakage of less than 0.01 volt when charged to 20 volts for 10 days. A 0.1 mg radium source was used to expose the pencils to about 4 mR in a 16-hour period. The pencils were read on a Hanford Ionization Chamber Pulse Reader and gave a calibration factor of 10.88 ± 2.37 mR/scale unit (full scale reading is 1.0). The TLD dosimeters were calibrated with the same source and corrected to background dose measured in another part of the building. Two overnight calibration trials gave factors of 0.018 ± 0.002 and 0.020 ± 0.001 mR per TLD reading. A longer calibration trial extending over a weekend gave a factor of 0.020 ± 0.003 mR per TLD reading. The sodium iodide crystal was also calibrated in the radiation field of the radium source and gave a calibration factor of 1.56×10^{-6} mR/hr per c/m with a background reading of 1.02×10^4 c/m.

Average population dose values for 1968 were computed from the diet estimates provided by adults at the Hanford

whole-body counter and the concentration of radionuclides in local foodstuffs for 1968. These dose estimates were corrected for the individual's weight in the case of bone and total body dose. The consumption levels were calculated from estimated frequency of consumption and the average adult meal size determined in a recent survey of serving sizes. A total of 6857 Tri-City adult diets was averaged. The average whole-body dose was found to be 2.3 mrem, of which 1.0 mrem was estimated external dose. The GI tract dose average was 19.2 mrem, also including 1.0 mrem external dose. The average thyroid dose was 16.1 mrem including 1.0 mrem external dose. Bone-seeking nuclides were found to total 0.5% of the intake limit.

Waterfowl Census

(R. F. Foster, R. E. Nakatani, and T. P. O'Farrell,
Environmental and Life Sciences Division)

Population Dynamics of Hanford Wildfowl

(Wayne C. Hanson, Terrestrial Ecology Section)

Considerable consultation was held with Environmental Evaluation personnel and Douglas United Nuclear engineers concerning the waterfowl contamination problem at the 100-K and 100-N Areas waste water trenches. Oral recommendations for biological control and sampling were tendered.

Preparation was made for the 1970 Canada Goose Nesting Survey. A major impact upon the population by flooding of nests had been anticipated in connection with the proposed Corps of Engineer's River Manipulation to be carried out in early April. The problem was averted.

Thermal Effects Study

(W. L. Templeton, Ecosystems Department)

AEC-1, Performance of Thermally Shocked Young Salmonids

(M. J. Schneider, Aquatic Ecology Section)

Differential predation tests to determine the escape performance of juvenile rainbow trout Salmo Gairdnerii continued. Juvenile fish were exposed to 23°C for 50, 25, and

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12 min prior to predation. No recovery period was allowed in any of the tests. Fish exposed for 50 min were preyed upon more heavily than controls. Exposure times of 25 and 12 min resulted in less obvious differences in relative predation rates. There appears to be an inverse relationship between temperature and exposure times to a temperature with regard to escape abilities.

Additional predator fish were marked and acclimated to 15°C. These fish were subjected to thermal resistance tests (30, 29, 28, 26, and 25°C) to determine the EL₅₀ and the LD₅₀ for comparison with fish used in earlier tests.

AEC-2, Gas Bubble Disease of Young Salmonids
(M. J. Schneider, Aquatic Ecology Section)

A surveillance of the gas saturation levels in the Columbia River was initiated this month. Water samples were taken above and below Priest Rapids dam, in the warm water seepage area near B-C Reactor site, in two slough areas and in the mainstem of the river at each of the last three locations. Since only one set of samples was taken, only general statements can be made about the river, sloughs and seepage area. The mainstem of the river, as well as the river above and below Priest Rapids Dam, is approximately air equilibrated. The water temperature is 3.5°C. The water in the sloughs is slightly less than saturated and is about 1.0°C higher than the mainstem.

In the laboratory, gas-supersaturated water was created thermally. Fish acclimated to air-equilibrated water at this higher temperature were exposed to the supersaturated water, and all symptoms of gas bubble disease were produced. The gas level in this tank was maintained at 120-135% nitrogen saturation and 110-125% oxygen saturation.

A single Doppler probe, made at this laboratory, was successfully implanted in three rainbow trout. A system was devised whereby round-the-clock monitoring of the fish's condition could be maintained. Output from the flowmeter was recorded on magnetic tape for 30 seconds every 5 minutes.

Preliminary results indicate that a) the first bubbles reach the heart 6-10 hr after the first exposure to chronic supersaturation, b) in early stages of gas bubble disease, the bubbles collect in the atrium and do not pass through the heart as long as the fish remains in a normal swimming position (since the atrium is the most dorsal portion of the heart, bubbles pass through in great bursts if the fish is rolled over), c) gas bubbles begin appearing on the skin about 72 hr or more after first exposure to chronic supersaturation, and d) when bubbles begin passing through the heart, the gills begin collecting gas as capillary occlusions. Pauley and Nakatani (1967) showed that other vital organs are also drastically affected by capillary occlusion.

AEC-6, Hanford Chinook Salmon Population Studies

(R. O. Gilbert, Applied Mathematics, Associated with Aquatic Ecology Section)

The study of the relationships between some of the environmental variables on the Columbia River and the estimated redd count over the period 1947-69 using stepwise regression techniques was completed. A report was prepared which describes the methods used, the variables investigated, the types of conclusions which can be drawn from such an analysis, the important conclusions, and recommendations for future and more exhaustive studies. A meaningful numerical representation of the influence of dam construction on redd counts was among hindrances in obtaining more meaningful results.

AEC-8, Effect of Temperature on Fish Disease

(M. P. Fujihara, Cellular and Molecular Biology Section)

River temperatures at White Bluffs remained at 40-41°F. The incidence of fish with columnaris antibodies, average titers, isolates and infection was 14%, 1:3, 0% and 0% respectively, similar to values observed last month. The percentage of fish with furunculosis antibodies increased slightly to 39%, but average titers (1:9) remained the same. Exposure (23%) and infection (4.5%) as determined by isolation methods continued to decrease.

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River temperatures at the Wenatchee site remained at 37°F; however, C. Columnaris was isolated from fish, despite continued cool temperatures. The percentage of fish with columnaris antibodies (47%) and average titers (1:23) was similar, but exposure (3.3%) was lower than last month. The incidence of fish with furunculosis antibodies (60%), average titers (1:18), exposure (17%) and infection (3.3%) were lower than observed last month.

Fish were also sampled approximately seven miles downstream from the Wenatchee site near a smelting plant on the Columbia River. Approximately 750-1000 gal/hr river water used as a coolant during the smelting process was being introduced into a cooling pond before release into the river. Water temperature at the release pipe was 75°F. The pond temperature was 70°F, but the pond was too shallow to set gill nets. Gill nets were set downstream from the smelting plant where temperatures ranged between 40° to 44°F. Studies were initiated to determine if fish residing in this warm water area of the river (3° to 7°F warmer than the Wenatchee site) will have a higher incidence of fish diseases than at cooler sites. Disease incidence at these sites will be compared with that at the White Bluffs and 100-K Reactor effluent seepage areas.

Columnaris isolates were not observed at the smelter site; however, the percentage of fish with antibodies (65.4%) and average titers (1:48) was higher than the Wenatchee site values of 46.7% antibodies and average titers of 1:23. Fish with furunculosis isolates (17.5%) and infection (1.8%) were similar to the Wenatchee site; however, the percentage of fish with antibodies (77.2%) and average titers (1:18) were higher than the Wenatchee site values of 60% and 1:14, respectively.

AEC-9, Effects of Increased Temperatures on Survival and Growth of Juvenile Steelhead

(P. A. Olson, Aquatic Ecology Section)

Juvenile steelhead exposed to approximate Columbia River temperatures and added increments of 3, 4 and 5°F since early

July have experienced yearly minimum temperatures during February and March near 42°F in the warmest lot. Mortalities have been inconsequential since last fall when most resulted from columnaris disease. Mortalities are not correlated with relative temperature conditions and range from a minimum of 4.0% in the 5°F increment to a maximum of 15.3% in the 3°F increment. Cold water temperatures continue to favor increased growth in the warmer lots so largest average fish are in the two warmer lots and smallest in the river temperature (coldest lot).

AEC-10, Temperature Tolerance of Aquatic Insects
(C. D. Becker, Aquatic Ecology Section)

Winter river temperatures increased only slightly, to 6.0°C, in mid-March. Ambient temperatures in 4 artificial channels receiving thermal increments at successive 2.5°C stages increased accordingly so that the warmest channel registered 16°C. In late February an abundant population of midge larvae (Diptera: Tendipedidae) appeared in the coldest channel (i.e., receiving undiluted river water) at temperatures of about 5.0°C. These larvae presumably issued from flights of ovipositing adult midges, which were observed two weeks previously with egg skeins attached. However, the greatest numbers of adults were associated with the channel heated to 10°C. The discrepancy between apparent preferred temperature for egg deposition and temperature under maximum appearance of midge larvae is not explainable at the present.

Taxonomic study of available midges continued. It was not possible to identify larvae due to the absence of reliable morphological criteria. Study of adults and pupae revealed that the majority collected in February and March belong to the subfamily Diamesinae, genus Diamesa. This complex group of midges reach greatest abundance in cold climates and swift streams, such as the Columbia River, but are little known in the Pacific Northwest.

Two TL₅₀ tests involving exposure of Diamesa larvae to temperatures of 5.0 (control), 7.5, 10.0, 12.5 and 15.0°C for 48 hr periods were completed. Mortalities over this range

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were insignificant, indicating that temperatures higher than 15°C are necessary to cause organism death from thermal shock. Larvae were noticeably more active at warmer test temperatures. Furthermore, a few more larvae developed into pupae during the relatively short observation period at the warmer temperatures.

Six troughs were constructed at the 100-F Area hatchery for rearing selected aquatic invertebrates in the laboratory under controlled temperature conditions. The additional troughs provide larger water volumes, which can be heat-regulated, for experiments and are suitable for TL50 tests.

Dosimetry Technology

(A. J. Haverfield, Environmental and Life Sciences Division)

Development of the Basic Dosimeter

All the punches in the die block for making the dosimeter carrying pouch were reground after the second encounter with misalignment between the die block and the selector rings. After this modification the performance of the perforator was very satisfactory for over 3,000 operations.

Studies continued to determine a quick anneal procedure for the Basic Dosimeter. An annealing temperature of 150°F has shown promising results over a three-week period. Studies are being made to assure that there is no fading with time.

A new heater unit was fabricated for the Basic Card Reader. The new design will permit greater tolerance in the X and Y direction; this will eliminate or greatly reduce the chance of the heater striking the plastic card.

The manual TLD card reader will transfer to the U. S. Testing Company March 30, 1970.

A newly designed heater, a new photomultiplier (PMT) assembly, an automatic delay and start circuit, and a visual and aural cycle indicator were installed. The high voltage

instability (less than 0.1%) was solved by correcting a 10 MHz oscillation in the power supply. The variation in dark current experienced in the field study should be eliminated with the new PMT assembly. This assembly was scheduled to go into the automated reader.

A descriptive booklet, specifically written for the manual TLD card reader, is being published since the converted Harshaw material was not deemed adequate to describe the operation and maintenance of the converted Harshaw TLD card reader.

Another point on the one-year annealing study was obtained 308 days after exposure on 5-8-69. This reading was only 3% below the reading of the first dosimeter in the study six days after exposure. The scatter in the reading over the 308-day period was + 2 and - 14 percent. This includes changes in reader conditions for the entire period. These results indicate only a slight fading, if any, over the 308 days.

Development of the Multipurpose Dosimeter

All the TLD-600's to be used in the prototype program were selectively chosen and marked to avoid confusion during assembly with the TLD-700's. Currently, the 700's are being selected by the exposure readout method to assure uniformity among the TLD to be used.

There will be a delay of four to five weeks in the delivery of the prototype multipurpose dosimeter because of procurement difficulties in obtaining the plastic. Study is being given to fabricating some units with an alternate material.

The Teflon tape for encasing the TLD ribbon in the multipurpose dosimeter was received and forwarded to the Mil-lor Company, the vendor for fabrication of the dosimeters.

Two selected photomultiplier tubes and focusing magnets were ordered. The latter, a new device, focuses only photoelectrons from the center portion of the photocathode into the

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multiplier window excluding extraneous electrons from the tube walls. This reduces the dark current by a factor of 5 to 10, which will help in the low dose determinations.

Several scope designs of the mechanized reader are being evaluated before starting the design of the mechanical handling system.

TLD's 100, 600 and 700 were exposed to neutrons and photons from PuO₂ (Yankee source) at the surface and at one inch. Positive neutron results were obtained after an eight-hour surface exposure and a twenty-hour exposure at one inch from the source. The maximum theoretical exposure calculated was 39 and 5 mrem/hr at the surface and at one inch, respectively. The TLD results are about half of the calculated values. Further work is scheduled during the forthcoming month.

Comparisons with tissue equivalent proportional counter data over the neutron energy range 0.48 to 1.4 MeV show that corrections can be made for the effects of moderating materials up to 1 1/2 in. thick with reasonable accuracy.

Exposures to beta radiation sources conducted previously with TLD-700 dosimeters showed some energy dependence in the response. For uranium beta radiation the response is about 0.38 count per mrad with a particular set of reader parameters. For ⁹⁰Sr beta radiation the response is about 0.54 count per mrad with the same set of reader parameters. These response numbers were obtained with only a thin paper covering the dosimeters.

This same type of TLD is to be used in the multipurpose dosimeter to measure penetrating and non-penetrating exposure. To achieve an energy response for the penetrating exposure which approximates a 1 cm depth dose curve, a filter composed of 0.025 in. of aluminum and 0.020 in. of plastic was developed. For uranium beta exposures of 1 rad the Al-plastic filter yields a response on the TLD of about 50 mrads. This response would be interpreted as penetrating exposure, which it is since the uranium source used for the exposure does emit some penetrating radiation. Other filter combinations

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tried did not yield a realistic result for the penetrating exposure when exposed to beta radiation.

The relative beta sensitivity of the TLD-700 dosimeters compared to Ra gamma radiation is 0.63 for ^{90}Sr and 0.45 for uranium beta. This means that an error in interpretation will be made in the case of mixed beta and low energy X-ray exposures. If a given non-penetrating result is interpreted as all X-ray when in fact it is entirely due to beta exposure, the error will be about 50% maximum. For a more realistic mixture the errors in interpretation should be no more than 20 to 30%. The penetrating dose measurement remains accurate as determined by the TLD filter combination.

A request for bid was sent out to several computer manufacturers to supply the small computer for the automated reader controller.

Environmental Dose Evaluation Concepts
(J. K. Soldat, Radiological Physics Section)

Re-evaluation was made of the factors recommended in January 1969 for calculation of doses to bone marrow and endosteal tissue from ^{32}P , ^{65}Zn and ^{90}Sr -Y incorporated into mineral bone. The originally suggested dose factors were based on the work of Dr. Spiers of the University of Leeds (United Kingdom). More recent published work by Spiers indicates that the factors estimated for ^{32}P were slightly conservative. Work reported by Dr. Spiers at the First IRPA Congress (Rome, September 1966) and the 2nd International Conference on Luminescence Dosimetry (Gatlinburg, Tenn., September 1969) indicates the new values for the dose factors for an adult which are tabulated in the following table. The original values estimated in January 1969 are also tabulated for comparison.

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DOSE FACTORS FOR ^{32}P IN ADULT SKELETON

<u>Item</u>	<u>New Estimates</u>	<u>Previous Estimates</u>
D_0 (a)	1.7 (d)	1.9 (e)
$\bar{\text{D}}_{\text{m}}/\text{D}_0$ (b)	0.2	0.3
$\bar{\text{D}}_{\text{s}}/\text{D}_0$ (c)	0.44	0.5

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- (a) D_0 - Mean skeletal dose to a small tissue-filled cavity - rads/hr per μCi contained in the skeleton.
- (b) $\bar{\text{D}}_{\text{m}}$ - Mean dose to bone marrow - rads/yr per μCi in skeleton.
- (c) $\bar{\text{D}}_{\text{s}}$ - Mean dose to endosteal tissue - rads/yr per μCi in skeleton.
- (d) Derived from the values given by Spiers for ^{90}Sr (2.7), ^{45}Ca (0.21), and ^{14}C (0.13).
- (e) This value is derivable from the ICRP recommendations (Publication II, 1959).

Since the estimated values do not include any contribution from ^{32}P incorporated in places other than the mineral bone (such as blood or red bone marrow itself), no change to the lower values is being recommended at this time. (Initial estimates of the potential additional dose from ^{32}P in these other places range from 10 to 30%).

The factors recommended in January 1969 for calculation of bone doses from intake rates are summarized in the following table.

DOSE FACTORS FOR CERTAIN BONE-SEEKING RADIONUCLIDES

<u>Nuclide</u>		<u>Adult</u> <u>(7 kg Skeleton)</u>	<u>Child</u> <u>(2 kg Skeleton)</u>
32p	Skeleton	0.19	0.66
	Bone Marrow	0.011	0.066
	Endosteal Tissue	0.019	0.066
65Zn	Skeleton	0.0031	0.011
	Bone Marrow	0.0028	0.010
	Endosteal Tissue	0.0028	0.010
90Sr	Skeleton	9.6**	9.6+ (34.)*
	Bone Marrow	0.39**	0.80+ (2.8)*
	Endosteal Tissue	0.81**	0.96+ (3.4)*

* Based on ICRP metabolic parameters being invariant with age unless otherwise noted.

** Approximately the same values can be derived from either the ICRP data at equilibrium or from specific activity.

+ Based on specific activity in the skeleton being 0.25 times that of the diet at all ages.

Radiation Protection

(H. V. Larson, Radiation Protection Department)

Promethium-147 was added to the list of radionuclides for which in vivo counting is useful. Phantom calibrations were performed for lung and head (bone) counting of ¹⁴⁷Pm, a low energy beta emitter. Bremsstrahlung radiation, which results from absorption of the beta particles, is detected with thin sodium iodide crystals giving minimum detectable limits of about 1.0 µCi and 0.2 µCi for the lung and head, respectively. Maximum permissible burdens for ¹⁴⁷Pm are about 12 µCi (lung) and 60 µCi (bone).

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Capability for in vivo counting of ^{90}Sr was improved by calibration of the lung counter for this radioisotope. Detection limit for an equilibrium mixture of ^{90}Sr - ^{90}Y is $0.018 \mu\text{Ci}$. Maximum permissible lung burden for ^{90}Sr is $0.8 \mu\text{Ci}$.

Use of a thermoluminescent dosimeter was implemented at the end of the month for approximately 4,000 Hanford employees whose duties are such as to require routine dosimeters for access rather than compliance (as defined by AECM Chapter 0524, expected to receive less than 10% of whole body dose limits). For economy and simplicity the dosimeter is a single lithium fluoride crystal suspended in a plastic card. Although exposure to mixed sources of radiation decreases the accuracy of the exposure measurement, by interpreting the dosimeter reading as a penetrating radiation dose, overall accuracy of $\pm 50\%$ is expected, with a detection level equal to or less than 50 millirads with adequate conservatism.

Evaluation of the basic thermoluminescent dosimeter is essentially complete. The following additional results were noticed:

1. Dosimeters with a previous radiation history of greater than 10 R cannot be reused with the recommended reader annealing procedure.
2. Temperature dependence was negligible below 100°F , and less than 15% over-response was noted at 120°F .
3. Storage at temperatures of 120°F for a five-day period had no effect on the reading, indicating that the dosimeters will be unaffected by the very hottest of Hanford summers.
4. No fading was noted over a one-month period and there seems to be no relationship to nor can fading be induced by changes in humidity or temperature of storage.
5. Light, both natural and artificial, can provide an extremely significant effect on the dosimeters, which could result in a spurious dose interpretation. The

manner of packaging the dosimeters apparently precludes any untoward effect. However, care should be taken to minimize errors from this source. Exposure to visible light should be limited to a few hours at most.

6. The reader was found to be extremely sensitive to ionizing radiation. Exposure levels of 0.1 mR/hr caused a doubling of the background counting rate of the reader and can inject errors of up to $\pm 50\%$ to the 100 mR level. Obviously, the reader must not be used in areas of fluctuating ambient radiation levels.

The Food and Drug Administration requested the AEC Division of Biology and Medicine to furnish current information on diethylenetriaminepentaacetic acid (DTPA), still classed as an experimental drug for the treatment of radionuclide deposition cases. At the request of HEHF, the apparent effectiveness of DTPA treatment for 19 Pu deposition cases for the period 1967 through 1969 was estimated and forwarded to HEHF for inclusion in a transmittal to DBM. The estimates, in some cases very approximate, indicated apparent body burden reductions of about 50% for wound injection cases and less than 10% for inhalation cases.

An integrated effort by ARHCO radiation protection representatives and BNW's Radiation Protection Department led to the initiation of a routine in vivo lung counting program for selected Pu workers. Examinations were scheduled to coincide with routine physical examinations for Pu workers. Detection of Pu from inhalation with this procedure is based on the measurement of low energy X-rays from ^{241}Am . Results of 10 cases to date indicated, from less than detectable ^{241}Am counts, that associated Pu lung burdens were less than 3 nanocuries, with the exception of one employee with a known previous small burden.

Arrangements were completed with U. S. Testing to provide analysis of bioassay samples for ^{249}Bk . Samples are being analyzed by a liquid scintillation technique which, although not as sensitive as desired, is the only current

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method of evaluation available pending procurement of an adequate source. Development is continuing on in vivo methods of detection.

Fifty-three special bioassay samples were scheduled and 31 in vivo examinations were conducted as a result of radiation occurrences. Special external exposure evaluations were performed for 6 Hanford contractor employees. There were no exposures in excess of the applicable limits. Among the radiation occurrences for whom special internal dosimetry evaluation was scheduled were several employees incurring nasal contamination up to 30,000 c/m from strontium storage tank blow-back into an operating area. In vivo examination indicated that the most highly exposed individual received a ^{90}Sr lung burden of less than 10% of the MPLB. As a result of another incident, several employees incurred particulate nasal contamination up to 5,000 c/m of activity later identified as ^{156}Eu by in vivo counting. The maximum deposition was less than 5% of the MPBB. A laboratory technician incurred high level ^{147}Pm skin contamination which resisted decontamination efforts. This employee was sent home with 30,000 c/m remaining on the tip of the finger. Although subsequent decontamination was effective, the dose to the thumb tip was estimated as 10-15 rem to a localized skin area.

Effective April 1, the new values for the standard PuF_4 and PuBe sources and the sigma pile go into effect. The new calibrations are based on recalculations of source output based on NBS calibrations and provide dose flux data in conformance with National Bureau of Standards Handbook 63.

The standard radium film jig was moved from the Annex Room into the Low Scatter South Room and modified to permit greater flexibility of use. Recalibration was accomplished with a minimum of trouble and indicates the source output is accurate to within the limits cited by the National Bureau of Standards.

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Environmental Evaluations

(W. L. Fisher, Environmental Evaluations Section)

The sampling of waterfowl in the 100-N and 100-K Areas produced another unusually radioactive duck. On March 6, 1970 (elsewhere erroneously reported as March 5), a mallard drake was shot at the 100-N trench. Radiochemical analysis indicated a ^{32}P concentration in flesh of 0.14 microcurie per gram, slightly more than the concentration (0.11 $\mu\text{Ci/g}$) reported last month to have been found in a duck from the 100-K trench. This bird also contained several other radio-nuclides in much smaller concentrations than the ^{32}P , as did the 100-K trench duck. The finding of these ducks prompted thorough surveys of 100-K and 100-N trenches. On March 7 trench bottom dose rate profiles were measured, and mud, algae and vegetation samples were obtained. Preliminary analyses indicated K and N trench inventories of at least 4 and 2 KCi , respectively.

The special waterfowl sampling was extended on March 13 to the 200 and 300 Areas. Between March 13 and March 29, 54 waterfowl were obtained from the 200 Areas and 2 from the 300 Area. Analytical results are just now becoming available. Nothing can be said at this point about the radio-activity of these birds, except that external contamination was detected on birds from B, U, Redox, and Honey Hill Swamps since the B-Plant strontium release to B Swamp. A goldfish was obtained from Gable Swamp on March 27.

Due to recent emphasis on waterfowl, no significant progress was made during March on the analysis of the Columbia River upstream from 100-F. It is hoped that the Environmental Evaluation effort can be redirected very soon in order to get back to the "opening the river" project.

Regional fallout levels increased to approximately 0.2 pCi/m^3 beta from 0.1 pCi/m^3 last month. This is approximately the same concentration as occurred during the same period one year ago.

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In general environmental levels of radioactivity were consistent with previous experience and indicated no unusual sources of exposure to the population or environs.

Meteorological Services
(E. H. Phinney, Synoptic Meteorology Section)

Meteorological services, viz., weather forecasts and observations and climatological services were provided to plant operations and management on a routine basis.

MARCH 1970
Forecasting

<u>Type</u>	<u>No. Made</u>	<u>% Reliability</u>
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General	62	86.0
Special	172	87.8

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